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13. ABSTRACT (Maximum 200 words)
The research and efforts mentioned above describe our accomplishments and new findings in three areas: (a) fundamental R/D; (b) new technologies and techniques; and (c) educational and training of research associates at Caltech. In the area of fundamental R/D, the laboratories are considered one of the national/international resources for the studies of the fundamental dynamics of chemical reactivity. Detailed description of the works are given in the publications in this report. As for the development of new technologies and techniques, our efforts, supported by the AFOSR, have been central to the development of ultrafast laser and electron technologies and their applications. These developments are essential to high-speed technologies, and, as mentioned in the previous report, Japan's MITI announced recently that Femtoscience and Femtotechnology are the key to many future industries of new materials, optical communication, switching,...etc.; they committed \$200 millions to this science and technology. Finally, the caltech group currently has close to 25 visiting associates, post-doctoral fellows, graduate students and undergraduate students.. The training and education of new generation of scientists and technologists in these areas have been extremely profitable, as evident by the leading positions these associates take in academic institutions and in the industrial sector.

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"Ultrafast Dynamics of Chemical Reactions"
December 15, 1993 - December 14, 1996

Progress Report

**+
Final Report**

August 12, 1996

20 AUG 1996

2. Objectives

The objectives of this research are central to the studies of the dynamics and mechanisms of chemical reactions, simple and complex. At Caltech our focus is on: (a) dynamics of elementary, complex, and atmospheric reactions; (b) experimental methodology for chemical control of reactions yield and channel; (c) reactions of high-energy states and of bimolecular type; (d) direct imaging techniques for ultrafast changes of structures. In these studies, the basic approach is based on the development of ultrafast laser (and electron) and molecular beam techniques to examine the nature of the dynamics and structures on the femtosecond time scale. Our goal is to relate these dynamics to features of bonding, and to possible control in the course of the reaction. Theoretical studies in our group and in collaboration with colleagues elsewhere are an important part of this research.

3. Status of Effort

Significant progress has been made in several areas of studies. These include (a) the development of new techniques for the studies of dynamics with *atomic-scale* resolution, (b) the applications to elementary, complex and atmospheric reactions, (c) the investigation of high-energy (Rydberg state) and bimolecular reactions, (d) the control of elementary reactions with femtosecond timing, and (e) the development of the new methodology of ultrafast imaging using electron diffraction and high power femtosecond lasers. So far the group has published some 40 papers.

4. Accomplishments/New Findings

The research and efforts mentioned above describe our accomplishments and new findings in three areas: (a) fundamental R/D; (b) new technologies and techniques; and (c) educational and training of research associates at Caltech. In the area of fundamental R/D, the laboratories are considered one of the national/international resources for the studies of the fundamental dynamics of chemical reactivity. Detailed description of the works are given in the publications in this report. As for the development of new technologies and techniques, our efforts, supported by the AFOSR, have been central to the development of ultrafast laser and electron technologies and their applications. These developments are essential to high-speed technologies, and, as mentioned in the previous report, Japan's MITI announced recently that Femtosience and Femtotechnology are the key to many future industries of new materials, optical communication, switching, ...etc.; they committed \$200 millions to this science and technology. Finally, the Caltech group currently has close to 25 visiting associates, post-doctoral fellows, graduate students and undergraduate students. The training and education of new generation of scientists and technologists in these areas have been extremely profitable, as evident by the leading positions these associates take in academic institutions and in the industrial sector.

5. Personnel Supported

The following graduate students and post-doctoral research fellows have been involved (supported, partially supported, or having their own fellowships) in this research: Dr. Jennifer Herek (presently at Lund University), Dr. Soren Pedersen (presently with McKinsey), Dr. R. Bowman (presently on the faculty at the University of Kansas), Dr. M. Gruebele (currently on the faculty at the University of Illinois), Dr. L. Khundkar (currently on the faculty at Northeastern), Dr. L. Bañares (currently on the faculty at Complutense de Madrid University), Dr. G. Roberts (at the University of Cambridge), Dr. M. Janssen (at the University of Amsterdam), Dr. P. Cong (on the faculty at the University of Hong Kong), Dr. A. Mokhtari (at Ecole Polytechnique), Dr. M. Rosker (at Rockwell), and Dr. T. Rose (Aerospace Corporation). At Caltech at the present time are graduate students S. Ahmed, S. Chong, J. Guo, M. Gupta, B. Horn, H. Ihee, Z. H. Kim, Q. Liu, C. Williamson, D. Zhang, and D. Zhong; post-doctoral research fellows S. Baskin, J. Cheng, and C. Wan; and post-doctoral scholar J. Cao. In addition, we had some collaborative efforts with Professor R. Marcus (Caltech), Professor K. Wilson (UC, San Diego), and Professor J. Polanyi (Toronto).

6. Publications (1995/1996)

Books

Collected Works (up to 1994)

Femtochemistry – Ultrafast Dynamics of the Chemical Bond, Vols. I and II,
World Scientific, New Jersey, Singapore

Articles

Femtosecond Real-Time Probing of Reactions: XVII Centrifugal Effects
in Direct Dissociation Reactions

G. Roberts and A. H. Zewail

J. Phys. Chem. Special Issue to Dr. Stuart A. Rice, **99**, 2520 (1995)

Dynamics of Ground-State Bimolecular Reactions

C. Wittig and A. H. Zewail

in: *Chemical Reactions in Clusters*, ed. E. R. Bernstein,
(Oxford, 1995) - Book Chapter, p. 64

Coherence -- A Powerful Concept in the Studies of Structures
and Dynamics

A. H. Zewail

Laser Physics **5**, 417 (1995)

Direct Observation of The Transition State

J. C. Polanyi and A. H. Zewail

Accounts of Chemical Research (Holy-Grail Special Issue),
28, 119 (1995)

Kinetic-Energy, Femtosecond Resolved Reaction Dynamics: Modes of
Dissociation (in Iodobenzene) from Time-Velocity Correlations

P. Y. Cheng, D. Zhong, and A. H. Zewail

Chem. Phys. Lett. **237**, 399 (1995)

Femtochemistry of Organometallics: Dynamics of Metal-Metal and
Metal-Ligand Bond Cleavage in $M_2(CO)_{10}$

S. K. Kim, S. Pedersen, and A. H. Zewail

Chem. Phys. Lett. **233**, 500 (1995)

Femtosecond Dynamics of Reactions: Elementary Processes of Controlled Solvation

A. H. Zewail

Berichte der Bunsengesellschaft für Phys. Chem.
99, 474 (1995)

Direct Femtosecond Observation of the Transient Intermediate in the α -Cleavage Reaction of $(\text{CH}_3)_2\text{CO}$ to $2\text{CH}_3 + \text{CO}$: Resolving the Issue of Concertedness

Sang Kyu Kim, Soren Pedersen, and A. H. Zewail
J. Chem. Phys. 103, 477 (1995)

Transition States of Charge-Transfer Reactions: Femtosecond Dynamics and the Concept of Harpooning in the Bimolecular Reaction of Benzene with Iodine

P. Y. Cheng, D. Zhong, and A. H. Zewail
J. Chem. Phys. 103, 5153 (1995)

Femtosecond, Velocity-Gating of Complex Structures in Solvent Cages

P. Y. Cheng, D. Zhong, and A. H. Zewail
J. Phys. Chem., submitted for publication

Femtochemistry and Max Bodenstein's Impact

A. H. Zewail

Springer Series in Chemical Physics - "Bodenstein Issue", In press

Femtosecond Molecular Dynamics of Tautomerization in Model Base Pairs

A. Douhal, S. K. Kim, and A. H. Zewail
Nature 378, 260 (1995)

Femtosecond Real-Time Probing of Reactions. XVIII: Experimental and Theoretical Mapping of Trajectories and Potentials in the NaI Dissociation Reaction

P. Cong, G. Roberts, J. L. Herek, A. Mohktari, and A. H. Zewail
J. Phys. Chem: Special Issue for Professor James L. Kinsey,
submitted for publication

Femtochemistry -- Advances Over a Decade

A. H. Zewail

Femtochemistry, M. Chergui (Ed.), World Scientific, Singapore (1996), p. 3

- Femtosecond Control of an Elementary Unimolecular Reaction from the Transition-State Region
A. Materny, P. Cong, J. L. Herek, and A. H. Zewail
Femtochemistry, M. Chergui (Ed.), *World Scientific*, Singapore (1996), p. 356
- Femtosecond Elementary Dynamics of Transition States and Asymmetric α -Cleavage in Norrish Reactions
S. K. Kim and A. H. Zewail
Chem. Phys. Lett. 250, 279 (1996)
- Proton-transfer Reaction Dynamics
A. Douhal, F. Lahmani, and A. H. Zewail
Chem. Phys. 207, 477 (1996)
- Femtosecond Real-Time Probing of Reactions: XIX Nonlinear (DFWM) Techniques for Probing Transition States of Uni- and Bi-molecular Reactions
M. Motzkus, S. Pedersen, and A. H. Zewail
J. Phys. Chem. 100, 5620 (1996)
- Femtosecond Chemically-Activated Reactions: Concept of Non-statistical Activation at High Thermal Energies
S. K. Kim, J. Guo, J. S. Baskin, and A. H. Zewail
J. Phys. Chem.
Accepted for publication
- Femtochemistry: Recent Progress in Studies of Dynamics and Control of Reactions and Their Transition States
A. H. Zewail
J. Phys. Chem., Centennial Issue, 100, 12701 (1996)
- Femtochemistry: Chemical Reaction Dynamics and Their Control
A. H. Zewail
Advances in Chemical Physics, In press
- Femtosecond Real-Time Probing of Reactions: XX. Dynamics of Twisting, Alignment, and IVR in the *trans*-Stilbene Isomerization Reaction
J. S. Baskin, L. Bañares, S. Pedersen, and A. H. Zewail
J. Phys. Chem., R. M. Hochstrasser Special Issue, 100, 11920 (1996)
- Femtochemistry of ICN in Liquids: Dynamics of Dissociation, Recombination and Abstraction
Chaozhi Wan, Manish Gupta, and A. H. Zewail
Chem. Phys. Lett. 256, 279 (1996)

Femtosecond Real-Time Probing of Reactions: XXI Direct Observation of Transition-State Dynamics and Structure in Charge-Transfer Reactions

P. Y. Cheng, D. Zhong, and A. H. Zewail

J. Chem. Phys.

Accepted for publication

Femtosecond Real-Time Probing of Reactions: XXII Kinetic Description of Probe Absorption, Fluorescence, Depletion and Mass Spectrometry

S. Pedersen and A. H. Zewail

Molecular Physics , In press

7. Interactions/Transitions

(a) Participation/Presentations at Meetings and Conferences

American Physical Society, Receipt of 1995 Herbert T. Broida Prize, "Atomic and Molecular Dynamics at Femtosecond Resolution"; (San Jose, California) March 20, 1995

University of Michigan, Moses Gomberg Lecture; "Chemistry at Femtosecond Resolution" (Ann Arbor, Michigan) March 27, 1995

University of Pittsburgh, 40th Annual Francis Clifford Phillips/Phi Lambda Upsilon Lectures; "Chemistry and Biology in the Femtosecond Age", "Future Directions in Femtochemistry"; (Pittsburgh, Pennsylvania) March 29 and 30, 1995

National Research Council of Canada Physical and Life Sciences Distinguished Lecture; "Atoms and Molecules in the Femtosecond Age"; (Ottawa, Ontario, Canada) May 2, 1995

Wesleyan University, 23rd Peter A. Leermakers Symposium; "Chemistry and Biology in the Femtosecond Age"; (Middletown, Connecticut) May 4, 1995

University of Heidelberg, 100 Years After Max Born Conference, Opening Keynote Lecture; "Recent Advances in Femtochemistry--Born's Impact"; (Heidelberg, Germany) July 25-28, 1995

Imperial College, 17th International Conference on Photochemistry, Plenary Lecture; "Femtosecond Photochemistry"; (London, England) July 30, 1995

University of Lausanne, Femtochemistry: The Lausanne Conference, Keynote Speaker; "Recent Developments in Femtochemistry"; (Lausanne, Switzerland) September 4, 1995

Yale University, The Inaugural Jerome A. Berson Lecture; "Chemistry and Biology at Femtosecond Resolution"; (New Haven, Connecticut) October 19, 1995

University of Groningen, Hendrik de Waard Lecture; "Femtoscropy, Seeing at the Speed of Light", (Groningen, The Netherlands) November 23, 1995

XXth Solvay Conference on Chemistry (Brussels, Belgium, November 28 - December 2, 1995); Opening Lecture, "Femtochemistry": Chemical Reaction Dynamics and Their Control", November 28, 1995; presentation to King Albert II at the Castle of Laeken, "Femtochemistry", November 29, 1995

Collège de France Lecture Series; "Chemistry and Biology at Atomic Time Resolution", (L'Institut de Biologie Moléculaire et Cellulaire, Strasbourg, France) November 21, 1995; "Femtochemistry in the Simple and Complex", (L'Institut de Physique et Chimie des Matériaux, Strasbourg, France) November 22, 1995; "Ultrafast Phenomena -- from Photography to Lasers", (Institut Henri Poincaré, Paris, France) December 5, 1995; "Future Directions for Probing and Controlling Matter", (Ecole Normale Supérieure, Paris, France) December 7, 1995

University of New Orleans, D. G. Davis Memorial Lecture, "Chemistry and Biology at Femtosecond Resolution", (New Orleans, Louisiana) March 22, 1996

American Chemical Society National Meeting (New Orleans, Louisiana): "Direct Observation of the Transition State -- 60 Years of Theory and Experiment", March 24, 1996; "Molecular Dynamics at Femtosecond Resolution", Peter Debye Lecture, March 26, 1996

Sixth Annual Richard B. Bernstein Memorial Lecture, "Femtochemistry -- Dick Bernstein's Major Influence and Vision", (University of California at Los Angeles, California), May 20, 1996

Ultrafast Phenomena (Tenth) Conference, "Femtosecond Chemistry", (Coronado, California), June 1, 1996

United States Air Force Office of Scientific Research Contractor Meetings, "Recent Progress in Studies of Femtosecond Dynamics and Control of Reactions", (Boulder, Colorado) June 4, 1996

ISSPIC 8: Symposium on Small Particles and Inorganic Clusters - Opening Lecture, "Femtodynamics of Clusters -- Atoms, Molecules, and Biological Systems", (Copenhagen, Denmark) July 1, 1996

6th International Royal Society of Chemistry Reactions Mechanisms Meeting, "Femtochemistry", (University of Kent at Canterbury) July 9, 1996

MOLEC 11th European Conference on Dynamics of Molecular Collisions, "Femtochemistry and Molecular Dynamics", (Denmark) September 1, 1996

Nobel Symposium, Femtochemistry and Femtobiology: Ultrafast Reaction Dynamics at Atomic-Scale Resolution, "Femtochemistry", (Björkborn, Sweden) September 9, 1996

A Vision for Chemistry: Frontiers Across Boundaries, British Association for the Advancement of Science, Annual Festival of Science, "Femtochemistry--Chemists End of the Time Race", (Birmingham, England) September 13, 1996

(b) Consultative and Advisory Functions

Advisory Editorial Board, *Chemical Physics*

Editorial Board of Advisors, *Laser Focus World*

Advisory Board, *American Men and Women of Science*

Advisory Board, *World Scientific*

Advisory Board of the Laser Facility, University of Pennsylvania

Advisory Board, King Faisal International Prize

Member of many national and international committees, conferences, etc.

(c) Transitions

Throughout the development of ultrafast imaging techniques we have exchanged the new findings on the bombardment of CCD's with electrons with the Jet Propulsion Laboratory, where these CCD technologies are an essential part of the Space Program. We have also benefited greatly from their expertise in this area. Contact person: Chief Technologist, Dr. Terry Cole.

Another collaboration we have is with the Aerospace Co. for the studies of reaction dynamics under unusual conditions of cluster formation and at low temperatures. Contact person: Dr. Jack Syage.

8. New Discoveries, Inventions, or Patent Disclosures

Recently, we have made one significant invention and two new discoveries. The invention involves the generation of ultrashort electron pulses in combination with a molecular beam apparatus. As mentioned in the previous report, it made it to the cover of the Journal of Physical Chemistry [J. C. Williamson and A. H. Zewail, Ultrafast Electron Diffraction. IV. Molecular Structures and Coherent Dynamics, *J. Phys. Chem.* **98**, 2766 (1994); M. Dantus, S. B. Kim, J. C. Williamson, and A. H. Zewail, Ultrafast Electron Diffraction. V. Experimental Time Resolution and Applications, *J. Phys. Chem.* **98**, 2782 (1994)]. This year we have observed the first real-time studies of the change of structures and we are in the process of publishing the work. We have also made (1) the first observation of electron motion crucial in molecular reaction phenomena (P. Y. Cheng, D. Zhong, and A. H. Zewail, Transition States of Charge-Transfer Reactions: Femtosecond Dynamics and the Concept of Harpooning in the Bimolecular Reaction of Benzene with Iodine, *J. Chem. Phys.* **103**, 5153 (1995), and (2) the first development of femtosecond nonlinear techniques to probe reactions under severe conditions of low concentration and high pressures (M. Motzkus, S. Pedersen, and A. H. Zewail, Femtosecond Real-Time Probing of Reactions: XIX Nonlinear (DFWM) Techniques for Probing Transition States of Uni- and Bi-molecular Reactions, *J. Phys. Chem.* **100**, 5620 (1996).

9. Honors/Awards

Wolf Prize in Chemistry, Israel (1993)
Carl Zeiss International Award, Germany (1992)
King Faisal International Prize in Science (1989)
First Linus Pauling Chair, Caltech (1990)
Elected to National Academy of Sciences (1989)
Elected to American Academy of Arts and Sciences (1993)
Elected to Académie Européenne des Sciences, des Arts et des Lettres (1994)
Fellow, American Physical Society

Earle K. Plyler Prize, American Physical Society (1993)
Nobel Foundation Lecture Series, Sweden (1994)
Bonner Chemiepreis, Germany (1994)
Herbert P. Broida Prize, American Physical Society (1995)
Leonardo Da Vinci Award of Excellence, France (1995)
Peter Debye Award, American Chemical Society (1996)
National Academy of Sciences, Chemical Science Award (1996)

M.A., h.c., (honorary degree) Oxford University
D.Sc., h.c., (honorary degree) American University
D.Sc., h.c., (honorary degree) Katholieke University (Belgium)
D.Sc., h.c., (honorary degree) University of Pennsylvania

Member:

National Academy of Sciences
American Academy of Sciences
European Academy of Arts, Sciences, and Humanities
American Physical Society
American Chemical Society
International Society of Magnetic Resonance
Inter-American-Photochemical Society
Society of Photo-Optical Instrumentation Engineers
European Photochemistry Association
Sigma Xi Society